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EUTROPHICATION AND WATER POLLUTION

Edward J. Martin and Leon W. Weinberger
Research and Development, Federal Water Pollution Control Administration, U.S. Department of the Interior, Washington, D.C.

Abstract. The problem of accelerated eutrophication may be defined as the state of a body of water resulting from intentional or unintentional, natural or man-made modifications to the aquatic environment to the extent that the ecological system supports an imbalance in the biological production and creates a nuisance or interference with a water use. This paper presents a survey of the problem of accelerated eutrophication, including a definition of the mechanism of eutrophication and an evaluation of probable sources and nature of the pollutional loads responsible for eutrophication. The problem involves a consideration of the type and quantity of nutrient material, especially nitrogen and phosphorus, from the point of view of control and the frequency and intensity of algal blooms. Both quantitative and qualitative data can be used to indicate the scope of the problem.

Wastes resulting from the municipal, agricultural, and industrial uses of water contribute significant quantities of nutrient compounds which may promote eutrophication in the nation's waterways. An estimated 235 to 2,350 tons of phosphate are discharged daily to receiving waters from municipal waste sources in the United States alone. The maximum efficiencies of seven treatment processes for removing nitrogen and phosphorus are reviewed. Fifty per cent of total phosphorus and 80% of total nitrogen are removed by use of an activated-sludge process while 99+% of the phosphorus and nitrogen are removed with a treatment sequence using ion exchange. These data indicate that treatment capabilities are available to control the discharge of nutrient compounds from point sources of wastes and to treat them to almost any desired degree. Therefore, the treatment and control technology is currently in hand to moderate the problem of accelerated eutrophication.

INTRODUCTION

Surface lakes and streams are becoming more frequently invaded by nuisance growths of aquatic organisms. Consequently, the beneficial use and reuse of the nation's water resources for municipal, industrial, agricultural, and recreational purposes is being seriously impaired. Bodies of water contain a constantly changing biological system composed of many microscopic and macroscopic life forms. These species are interrelated and depend on the condition of the total ecological balance of the environment for their existences. The biological system is dynamic, is complex, involves biochemical interrelationships among the life forms on the bottom and life forms in the water itself, and involves chemical and hydrologic interactions between the bottom sediments and the water. Four factors that influence the aquatic ecological balance are: the concentration of suspended and dissolved organic and inorganic compounds, the availability of these compounds as nutrient material, the concentration of dissolved gases including oxygen, and the availability of sunlight.

Lakes and streams experience a "life cycle" in that they are born, they age, and may eventually become extinct. Characteristic species and numbers of life forms and environmental conditions are associated with each phase of the aging process. In general, formation (birth) of lakes is followed by a period, termed oligotrophy, that may be characterized by low temperatures, great depth, clear water, and low to moderate biological production. As the aging process continues, the body of water may become eutrophic, generally characterized by higher temperatures, decreased depth, and increased biological

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production. A body of water may even become dystrophic (old age) and even "die," such as becoming a peat bog and, ultimately, dry land. This aging process has been occurring naturally for eons.

Modifications of the ecological system, either natural or man-made, can produce changes which affect the whole system. Inorganic nutrients are necessary for biochemical synthesis of cell material. Consequently, the life processes depend on the quantity, nature, and rate of addition of nutrients. An increase in the availability of nutrient material may stimulate biological production and promote the premature aging of a lake, called eutrophication. In some instances, this may have desirable side effects. The most commonly recognized sign of advancing eutrophication is the algal "bloom." A bloom is the extensive growth of a single or limited number of species to such an extent that nuisance conditions develop and some water use is impaired. Nitrogen and phosphorus have been considered as contributing to nutrient fertilization and subsequently to algal blooms.

The entire process of the premature aging of water bodies—eutrophication—including the development of algal blooms, is not completely understood. At present, it is known that wastes resulting from man's use of the water contain nitrates, phosphates, and other nutrient materials. Undoubtedly, the addition of wastes containing nutrients may result in undesirable changes in a body of water. The extensive and rapid degradation of large lakes, such as Lake Erie, has prompted both public concern and increased research to solve the problem.

This paper presents a statement of the problem and its terminology, defines the mechanism of eutrophication, and enumerates the probable sources and characteristics of pollutional loads responsible for accelerated eutrophication. Available treatment processes to remove nutrients from wastewaters are reviewed.

STATEMENT OF THE PROBLEM

Nutrients

Significant among plant nutrient materials are inorganic compounds. Nitrogen and phosphorus, particularly as nitrates and phosphates (Sawyer and Buzzell 1962), seem to be the principal compounds involved in plant growth. These compounds are generally more readily available for use in plant metabolism when they are in solution. This is the case in many raw municipal and industrial waste effluents; secondary biological treatment processes tend to break down or modify waste components into a more usable form.

The prolific growth of algae and other aquatic vegetation, stimulated by large quantities of inorganic nutrients in waste effluents, has a number of significant corollary effects: 1) the death and decay of aquatic vegetation exerts a dissolved oxygen demand through biological degradation. Algae produce oxygen through photosynthesis and thereby increase the ability of lakes and streams to assimilate wastes. However, death and decay of large quantities of algae impose an additional load on the ecological system since oxygen is no longer produced by the algae but is required to degrade the algal cell material. 2) After degradation, the inorganic nutrients are released back into the water; this tends to make the process self-perpetuating. 3) Aquatic vegetation, particularly phytoplankton, is essential in the food chain, and an increase or decrease in the production of algae can induce subsequent imbalances in more highly developed species including fishes. The resulting imbalances could modify the ecological system to the extent that beneficial uses of water such as for recreation and commercial fishing are attenuated.

Besides the fundamental changes induced in the total environment, excessive algal growth imparts undesirable tastes and odors to water supplies (Sigworth 1957); releases toxins resulting in death of animals (Grant and Hughes 1953); and interferes with sandfiltering operations in water treatment plants (Palmer 1962). The toxin-producing algae include some forms of *Microcystis* and *Anabaena*.

Not all changes in the aquatic environment produce deleterious effects. The growth of aquatic plants releases dissolved oxygen to the water, which permits the degradation of waste materials and assists in maintaining desirable aesthetic conditions. Waste stabilization ponds used to treat sewage are based on this principle. Also, a biologically underproductive lake poses no challenge to the sport fisherman, and an increase in eutrophication may increase the use of such a lake.

To maintain an ecological balance that enables the optimal use of water resources, it will be necessary to adopt criteria for nutrient concentrations and to apply nutrient control techniques to the nation's waterways.

Nutrient Concentrations

Nitrogen and phosphorus have been considered as essential ingredients for promoting the growth of aquatic vegetation. However, they may not be the "critical" nutrients or the nutrients that determine whether or not growth occurs. Extremely small concentrations of other elements—micronutrients—may be critical to the growth of aquatic vegetation under some conditions. Some studies have indicated that elements such as cobalt and manganese, in small quantities, are important in algal nutrition (Eyster 1964; Krauss 1961).

Figure 1 shows concentrations of total phosphorus (P) and nitrate-nitrogen (NO_3-N) in various waters and wastes. The values represent averages and

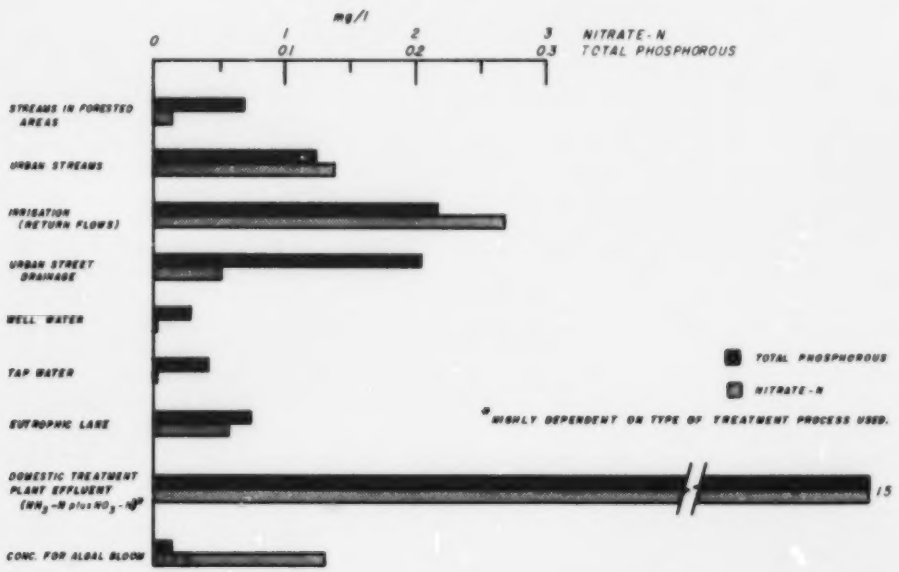

FIG. 1. Typical concentrations of nitrate-nitrogen and total phosphorus in some waters of interest. Values are averages and are to be used only for comparisons in a gross sense. Data are from various sources.

TABLE 1. Summary of water pollution surveillance system data.

Water year	Number of reporting stations	Stations where values equal or exceed 0.1 mg/1 PO_4		Stations where values equal or exceed 0.2 mg/1 PO_4	
		Number	Per cent	Number	Per cent
1961-62	116	99	85.3	66	56.9
1962-63	114	82	72.0	59	51.7
1963-64	115	84	73.0	65	56.5
Total	345	265		190	
Average			77.0		55.0

TABLE 2. Occurrence of high nitrate waters in U.S., water year 1962.*

Number of stations reporting nitrate concentrations higher than 1.4 mg/1		
	Surface water	Ground water
Alabama		1
Alaska	2	2
Arizona	1	2
Arkansas	1	2
California	2	2
Colorado	2	
Connecticut	1	1
Florida	9	1
Georgia		1
Iowa		2
Kansas	7	1
Maryland		1
Michigan		1
Minnesota	3	
Nebraska	4	1
Nevada		1
New Jersey		2
New Mexico	2	1
New York	1	1
North Carolina	3	1
North Dakota	4	2
Ohio	6	
Oklahoma	3	
Oregon	1	
Pennsylvania	9	
Rhode Island		1
South Dakota	2	
Texas	11	4
Utah	2	1
Washington	1	1
West Virginia		1
Wyoming	2	2
Washington, D.C.		1
Puerto Rico		1

*From unpublished data of S. K. Love, Water Resources Division, U.S. Geological Survey, Washington, D.C.

are to be used only for gross comparisons. "Concentrations for algal bloom" are to be interpreted only as the concentrations which are likely to precede nuisance bloom conditions if the concentrations occur or are exceeded during periods of low biological production (such as winter) and if climatological conditions are favorable for growth; they are not maximum values which should not be exceeded throughout the entire year. The values presented in Fig. 1 were obtained by analyzing data from various sources (Sylvester 1961; FWPCA 1960-1964[1]). Sawyer (1947, 1962) found that concentrations of 0.1 milligrams of soluble phosphorus (P) per liter (0.0326 as PO_4) and concentrations of 0.30 mg of inorganic nitrogen (N) per liter at the time of the spring overturn may be sufficient to support algal blooms in selected lakes during the summer. This could occur only if other environmental growth conditions are optimum. The value of 0.30 mg of inorganic nitrogen (N) per liter was converted to 1.4 mg of NO_3-N (nitrate-nitrogen) per liter for inclusion in Fig. 1. Between 1961 and 1963, the Water Pollution Surveillance System of the Public Health Service operated approximately 130 sampling stations on river systems throughout the United States. During this time the plankton populations were counted bi-weekly and phosphate concentrations were determined weekly. More than 49% of the stations counted an average of at least 10,000 algae per milliliter. This represents a concentration of organisms that would produce an algal bloom condition in the waters. A "bloom" has been defined as a

[1]Unpublished data, Research and Development, Federal Water Pollution Control Administration, U.S. Department of Interior, Washington, D.C.

water having about 500 individuals of a given species of phytoplankton per milliliter (Lackey 1945). However, if a bloom is considered as a condition which interferes with a water use, phytoplankton concentrations as low as 10 standard units per milliliter have imparted a noticeable taste to water supplies (Symons 1956).

The fertilization of rivers is further indicated by the phosphate content of samples collected from the stations referred to above. A summary of phosphate concentrations is presented in Table 1. An average of 77% of the stations reported that the water samples contained at least 0.1 mg PO_4/l, while an average of 55% of the stations reported at least 0.2 mg/l.

The nitrate contents of surface and ground waters are determined by the U.S. Geological Survey on a nationwide basis. Table 2 summarizes the number of stations in various states which reported nitrate concentrations greater than 1.4 mg/l during the 1962 water year. These data, both for phosphorus and nitrogen, indicate that high concentrations of these nutrients are present throughout the country. Consequently, the potentiality for algal blooms is widespread.

It is useful to consider not only the waste concentrations in waterways but also the total quantity of nutrients involved in waste discharges. The Illinois River Waterway, including treated sewage effluent from treatment plants in Chicago, was studied by Hurwitz, Beaudoin and Walters (1965) and illustrates this point. Three treatment plants in Chicago reported that as high as 76.7% of the phosphates were removed from the effluent. Even though this percentage is high, more than 32 tons of phosphate are added to the Illinois waterway system per day (Table 3). The amount added by tributary flows is about 11 tons per day, totaling about 43 tons per day. These calculations indicate that even if the percent removal of phosphate is increased through modifications to existing treatment processes, a large amount of nutrient material as phosphate would still be added to the nation's surface and ground waters.

Control techniques must be developed and used to reduce the concentrations of nitrogen and phosphorus in waste effluents and surface water while research proceeds to elucidate the detailed biochemistry of algal nutrition for still more effective and economic control.

TABLE 3. Average phosphate added to Illinois waterway. June-September 1960. Data from Hurwitz et al. (1965).

	Flow rate (cfs)	Total PO_4 in effluent (mg/l)	Phosphate removal efficiency	Tons of phosphate added daily
Chicago Area Treatment Plants				
Northside	363	12.8	36.6	12.55
Southwest	1475	3.75	76.7	14.95
Calumet	211	8.38	53.9	4.77
Subtotal				32.27
Tributaries				
Des Plaines R.	320	1.76	—	1.52
Du Page R.	115	2.29	—	0.71
Kankakee R.	1700	0.56	—	2.57
Fox R.	1300	1.47	—	5.29
Vermillion R.	220	1.57	—	0.93
Subtotal				11.02
Total tons per day				43.29

Algal Blooms

Changes in the frequency and intensity of algal blooms, even though qualitative, may indicate accelerated eutrophication. Selected surface waters in the United States, where the growth of algae has been sufficient to cause taste and odor problems and other deleterious effects, are listed in Table 4. Additional data on rivers during 1957 through 1960 (Table 5) illustrate that the problem of algal growth is not confined to local or regional areas. Significantly, most of these rivers contained greater than 1,000 organisms per milliliter during more than one-half of the time. The "Maximum Count" values in most cases are high. The most frequently occurring types of algae and their geographic area of occurrence are summarized in Table 6. One group of algae, *diatoms*, has been notoriously associated with the clogging of sand filters in water treatment plants throughout the country, besides causing problems of taste and odor associated with other algal forms.

TABLE 4. A partial list of eutrophic waters in the United States.

California	Sacramento Delta Estuary
Connecticut	Lake Zoar
Long Island	Marine water around Long Island
Maine	Lake Sebasticook
Maryland - Virginia	Potomac River estuary
Michigan	Shorelines of southwestern Lake Michigan
Minnesota	Shagawa Lake
Oregon	Klamath Lake
Ohio	Lake Erie
Washington	Lake Washington
Wisconsin	Madison Lakes

This problem of accelerated eutrophication is not confined to the United States. Some lakes in Germany and Switzerland are experiencing similar conditions (Hasler 1947).

Accelerated Eutrophication

During geologic time, lakes and streams have been undergoing a natural aging process, which induces hydrologic and ecological changes in the environment. The age classification of lakes as oligotrophic, mesotrophic, or eutrophic, depends on the degree of biological production, the types of species, and the environmental conditions, including nutrient and dissolved oxygen concentrations (Hutchinson 1957).

TABLE 5. Abundance of algae in American rivers, October 1957-September 1960. From Palmer (1964).

River	State	Sampling point	Counts above 1000/ml (%)	Maximum count
Mississippi	Iowa	Burlington	98	53,060
Arkansas	Oklahoma	Ponca City	78	101,200
Delaware	Pennsylvania	Philadelphia	75	4,750
Ohio	Ohio	Cincinnati	74	30,010
Missouri	Kansas	Kansas City	63	44,300
Potomac	Maryland	Great Falls	62	43,160
Red	Louisiana	Alexandria	61	29,070
Yellowstone	Montana	Sidney	61	21,480
Rio Grande	Texas	Laredo	51	34,510
Columbia	Washington	Pasco	47	4,370
Merrimack	Massachusetts	Lowell	41	16,250
Hudson	New York	Poughkeepsie	41	3,730
Colorado	Arizona	Yuma	37	5,050
Savannah	Georgia	Port Wentworth	28	10,860
Tennessee	Tennessee	Chattanooga	28	4,400

TABLE 6. Nuisance algae most frequently reported in American water supplies (Recorded by geographic regions). From Palmer (1964).

	North-eastern	North Central	South Central	South-western
Diatoms	x	x	x	x
Asterionella	x	x	x	-
Cyclotella	x	x	x	x
Fragilaria	x	x	x	x
Melosira	x	x	x	x
Stephanodiscus	-	x	-	x
Tabellaria	x	x	x	x
Blue-green algae				
Anabaena	-	x	x	x
Anacystis	x	x	x	-
Aphanizomenon	x	x	-	-
Coelosphaerium	x	x	-	-
Oscillatoria	x	x	x	x
Green algae—nonfilamentous				
Pediastrum	x	x	x	x
Scenedesmus	-	x	x	-
Staurastrum	x	x	-	x
Green algae—filamentous				
Chara	-	-	-	x
Cladophora	-	x	-	-
Pithophora	x	-	x	-
Spirogyra	x	x	x	-
Vaucheria	-	x	-	x
Flagellates—pigmented				
Ceratium	x	x	-	x
Chlamydomonas	x	x	x	-
Dinobryon	x	x	-	-
Euglena	-	x	-	-
Peridinium	-	x	-	-
Synura	x	x	-	-
Uroglenopsis	x	x	-	-

The problem of accelerated eutrophication may be defined as: the state of a body of water resulting from the intentional or unintentional, natural or man-made modifications to the aquatic environment to the extent that the ecological system supports an imbalance in the biological production; this creates a nuisance or interferes with a water use. The problem is of immediate concern because man's activities result in enormous quantities of wastes, which increase the rate of enrichment by many fold. Among these "activities" are: 1) discharge of inadequately treated sewage and industrial wastes, 2) improper agricultural and other watershed practices, and 3) canalization of streams.

A schematic representation of eutrophication is given in Fig. 2, in terms of nutrient inflow and outflow. When the inflow of nutrient material exceeds the outflow, nutrients can accumulate in the body of water. The accumulation is reflected in the generally increased biological production. The "fixed" bottom is that portion of the bottom of the water body which does not interact through physical, chemical, or biological processes with the aquatic environment.

Because the eutrophication process is very complex there are only a few analytical methodologies which yield consistent and usable results. Chemical analyses may be used to assess the degree of eutrophication, but the difficult

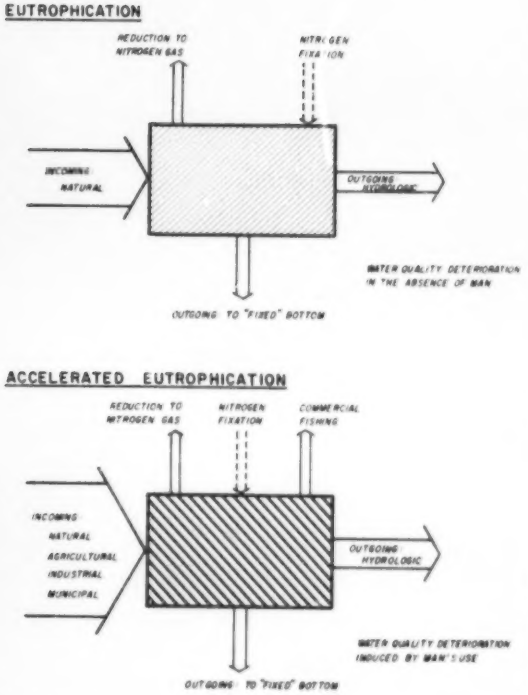

FIG. 2. Schematic diagram of eutrophication in the absence of man and the problem of accelerated eutrophication as induced by man's use of water. The various mechanisms by which nutrient material may enter and leave a lake are depicted.

decision is to determine which compounds to measure; this, in turn, depends on which compounds are critical nutrients. In general, eutrophic lakes contain blue-green algae and warm water fishes, are richer in nutrients, and support greater biological production than "clean water" lakes. However, a definition based on any one of these parameters may be inadequate. The quantitative determination of biological production is even less well defined. The development of appropriate analytical techniques is a pressing research need.

SOURCES OF NUTRIENTS

Nitrates and particularly phosphates are not analyzed routinely at municipal sewage treatment plants nor during the monitoring of industrial waste effluents. Only recently, the quantity of nitrogen and phosphorus compounds in wastes has been recognized by sanitary engineers as an important factor in inducing aquatic ecological changes. As a consequence, no detailed quantitative accounts of plant nutrients in various types of industrial wastes can be presented. Some data are available, however, to indicate the magnitude of the problem.

The relative amounts of water used as municipal, irrigation, industrial and power supplies, and totaling 300 billion gallons per day (BGD) in the United States, are shown in Fig. 3. The values do not represent waste quantity. To indicate the magnitude of nitrates and phosphates in water supplies, the quantities of these nutrients are calculated in terms of municipal waste discharges. In the United States, the total daily municipal waste discharge is 11.2 BGD; this value is calculated on the basis of a municipal-water use value of 14 BGD (Fig. 3) and a 20% consumptive use value (water which is used but not returned as waste). Phosphate concentrations

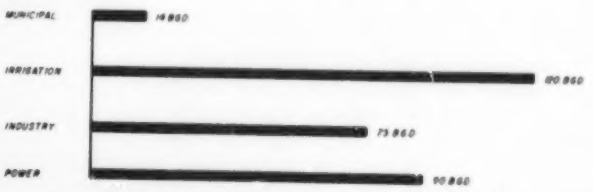

FIG. 3. Average estimated daily water use values for municipal, irrigation, industrial and power production uses in the United States. Total usage = 300 BGD. If the water use values for "Industry" and "Power" are added, the total of 165 BGD will exceed the use for "Irrigation." From Landsberg (1964).

in municipal secondary effluents range from about 5 to 50 mg PO_4/l of water (FWPCA, see footnote 1). Based on these figures, 235 tons to 2,350 tons of phosphate per day would be discharged to receiving waters around the country. These calculations are based on concentrations in municipal wastes which have received secondary biological treatment.

The percent of nutrient materials, particularly phosphates, removed from waste waters by secondary treatment plants varies greatly among plants. Phosphate removals have been reported as high as 77%; the average is probably about 10%. Total nitrogen removal by secondary treatment plants is about 50% (Stephan 1965b); the chemical form of nitrogen remaining in the effluent depends largely on the operating characteristics of the plant. Based on an average concentration of 15 mg of nitrate-nitrogen per liter of secondary effluent (Stephan 1965a), an average of about 700 tons of nitrate-nitrogen per day is present in secondary effluents in the U.S. A portion of the phosphates and nitrogen in municipal wastes is of human origin. A portion of the phosphates is from detergents.

The nutrient load resulting from agricultural and industrial water uses is extremely difficult to estimate. About 4.4 million tons of nitrogen (N) and 3.4 million tons of phosphorus (as P_2O_5) were included in fertilizer products manufactured during Fiscal Year 1964, (U.S. Dept. of Agriculture 1965); a fraction of this quantity reaches the nation's lakes and streams. It is not known what percentage of these nutrients leaches through the soil, enters surface waters, and contributes ultimately to the accelerated eutrophication problem. The quantity of nutrient compounds discharged to receiving waters and their subsequent effects on lakes and streams will vary regionally, depending on climatological conditions, types of water uses, and amount of dilution water available.

The nutrient content of wastes from other than municipal and agricultural sources is difficult to estimate because of their diverse compositions. Since more water is used for industrial purposes and power production than for agriculture on a nationwide basis, the nutrient contribution of industrial wastewaters should be further investigated.

TREATMENT PROCESSES FOR NUTRIENT REMOVAL

Several processes are available for removing nutrients, especially nitrogen and phosphorus compounds, from wastewaters. Eight of the most widely known processes are: 1) activated sludge, 2) algal removal systems, 3) chemical precipitation, 4) ion exchange, 5) ammonia stripping, 6) electrodialysis and other membrane processes, 7) effluent spraying on land, and 8) distillation.

It is useful to consider the nutrient removal efficiency of the various processes as well as the maximum removal efficiencies that can be obtained under various conditions. Stephan (1965b) reviewed the removal efficiencies of various components in treated wastewaters including total phosphate (PO_4) and total nitrogen (N) as well as the effects of superimposing various advanced waste treatment processes on conventional primary and secondary treatment processes. These data are presented in Table 7. The overall removal efficiencies reported in Table 7 are, for a given unit process, placed in series or in parallel with preceding unit processes.

Maximum achievable removal efficiencies for nitrogen and phosphorus compounds have been reported for both modified activated-sludge processes and for other processes (Table 8). These data indicate a lack of uniformity in reporting analytical results, particularly with regard to forms of nitrogen and phosphorus. Figure 4 summarizes the maximum expected removal efficiencies

TABLE 7. Estimated removal efficiencies for water renovation processes. Adapted from Stephen (1965b). Individual process removals } over-all removal (per cent removals based on raw waste concentration).

Process	Suspended solids (SS)	BOD (carbonaceous)	Chemical oxygen demand (COD)	Methylene blue active substance (apparent ABS)	Total phosphate	Total nitrogen	Total dissolved solids (TDS)
[C = conventional primary & secondary treatment] ↓ C ↓	90 } 90	90 } 90	80 } 80	50 } 50	10 } 10*	50 } 50	5 } 5
[A_1 = foam separation] ↓ C ↓ A_1 ↓	90 2 } 92	90 3 } 93	80 5 } 85	50 35 } 85	10 0 } 10	50 0 } 50	5 0 } 5
[A_2 = lime-alum coagulation & sedimentation & rapid sand filtration] ↓ C ↓ A_2 ↓	90 9 } 99	90 3 } 93	80 5 } 85	50 5 } 55	10 85 } 95	50 0 } 50	5 5 } 10
[A_3 = adsorption] ↓ C ↓ A_2 ↓ A_3 ↓	90 9 0 } 99	90 3 6 } 99	80 5 14 } 99	50 5 40 } 95	10 85 0 } 95	50 0 5 } 55	5 5 5 } 15
[A_4 = electrodialysis] ↓ C ↓ A_2 ↓ A_3 ↓ A_4 ↓	90 9 0 0 } 99	90 3 6 0 } 99	80 5 14 0 } 99	50 5 40 3 } 98	10 85 0 2 } 97	50 0 5 20 } 75	5 5 5 35 } 50
[A_5 = evaporation] ↓ C (50% → A_2 → A_3; 50% → A_5) ↓	90 (.5)9 (.5)0 } 99	90 (.5)3 (.5)6 } 99	80 (.5)5 (.5)14 } 99	50 (.5)19 (.5)40 } 97	10 (.5)85 (.5)0 } 96	50 (.5)0 (.5)5 } 75	5 (.5)5 (.5)5 } 55

*10% removal is a nominal minimum; variability among plants is great and removals of over 70% have been reported.

TABLE 8. Nutrient removal efficiencies of various treatment processes (Maximum achievable efficiencies unless the range is indicated).

Treatment process	Total[1] nitrogen removal (%)	Total[1] phosphorus removal (%)	Scale of operation	Reference
Biological processes	–	47.1 (PO_4)[2]	Full	Wirtz (1966)
Activated sludge	22-82	–	Lab	Culp and Slechta (1966)
	20-77	–	Lab	Ludzack and Ettinger (1962)
	–	80 (ortho PO_4)	Lab	Levin and Shapiro (1965)
	–	10-30 (ortho PO_4)	Full	Levin and Shapiro (1965)
	–	25-38 (ortho PO_4)	Lab	Feng (1962)
	–	37-77 (PO_4)	Full	Hurwitz et al. (1965)
	30.6-69.3	–	Full	Johnson (1958)
	63.3-69.2	–	Lab	Johnson and Schroepfer (1964)
Algae	95 (total inorganic)	70	Lab	Gates and Borchardt (1964)
	38.5 (increase) (PO_4)	41.3	Full	Rand and Nemerow (1964)
	–	44.4-95[3]	Lab	Rand and Nemerow (1965)
	35 (NH_3-N)	85 (PO_4)[4]	Lab	Fitzgerald and Rohlich (1964)
	65[5]	100[5]	Pilot	van Vuuren et al. (1965)
	–	90	Lab	Bogan (1961)
Activated sludge—chemical	–	95 (approx.)[6]	Lab	Tenney and Stumm (1965)
Chemical (lime)—algae	–	97 (approx.)	Lab	Sawyer and Buzzell (1962)
Chemical processes[7]				
Lime	–	93.5 (PO_4)	Lab	Rand and Nemerow (1965)
	–	99	Lab	Lea et al. (1954)
	12.1-24.4[8]	77.7-90.7[8]	Lab	Sawyer and Buzzell (1962)
	–	92-100 (PO_4)	Lab-Pilot	Bishop et al. (1965)
Alum-lime	–	95	Lab	Lea et al. (1954)
Alum	–	97	Lab	Rand and Nemerow (1965)
	60 (Org.-N)	94	Lab-Pilot	Lea et al. (1954)
	68 (Org.-N)	99 (Sol. PO_4)	Lab	Rohlich (1961)
	–	96-100 (PO_4)	Lab-Pilot	Bishop et al. (1965)
Iron	–	99 (Sol. PO_4)[9]	Lab	Rohlich (1961)
Other processes[10]				
Ion exchange	–	98.5-99.6	Lab	Rand and Nemerow (1965)
	84	95	Lab-Pilot	Eliassen et al. (1965)
	82-99.5 (NH_3-N)	–	Lab	Culp and Slechta (1966)
Ammonia stripping[11]	98 (NH_3-N)	–	Lab	Culp and Slechta (1966)
	82 (NH_3-N)	–	Lab	Nesselson (1954)
Electrodialysis	50	50 (PO_4)	Lab-Pilot	Stephan (1965b)
Effluent spraying on land[12]	54-68	76-93	Full	Foster and Ward (1965)

[1]Except where indicated.
[2]Average of monthly averages, December 1965 and February 1966.
[3]Cultivation time from 4 to 37 days.
[4]pH dependence reported.
[5]Flocculation and algae flotation.
[6]At optimum alum-polyelectrolyte dose.
[7]At optimum dosage and pH conditions.
[8]On raw sewage.
[9]Ferrous or ferric sulfate.
[10]In conjunction with pre-treatment or synthetic feed.
[11]pH dependent.
[12]Dependent on soil loading and climatological conditions.

by the use of various treatment processes. These values are the most likely estimates, based on this literature review.

Activated Sludge

The total amount of phosphate removed from wastes by the activated-sludge process depends on which processes and techniques are used to handle and dispose of the sludge. Variations in these methods may account for widely differing efficiencies among treatment plants. Other factors such as aeration rate, aeration time, aeration solids and return sludge ratios also may affect the percent of phosphate removed. The percent of phosphate removed at three treatment plants in Chicago varies from 37 to 78% (Table 3). Variations in these percentages may be due to differences in the types of wastes treated. Also, studies at the Baltimore, Md., and Washington, D.C. (Blue Plains) plants showed that the total phosphate content may increase between the influent and effluent of the activated-sludge secondary treatment plants (Levin 1963).

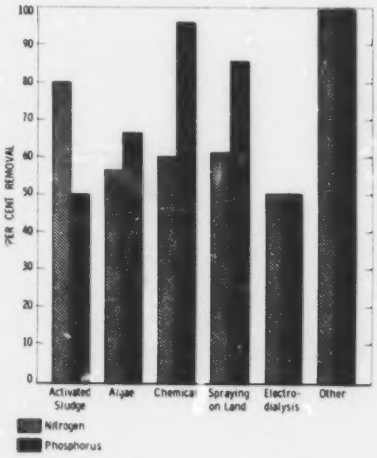

FIG. 4. Estimated maximum process removal efficiencies based on this literature review. Values for "Chemical," "Spraying on land," Electrodialysis" and "Other" are values for a process sequence which includes pretreatment, if necessary.

With increased aeration during the activated-sludge process, orthophosphate removal efficiencies were about 30% in a 60-million gal/day plant-scale demonstration and "occasionally" as high as 80% in controlled laboratory experiments (Levin 1963). He concludes that dissolved oxygen is essential to orthophosphate uptake which is attributable to oxidative phosphorylation. In normal sewage treatment plant operations, the dissolved, inorganic orthophosphate "leaks out" of sewage organisms when the dissolved oxygen level decreases. He also showed that a maximum rate of orthophosphate uptake occurred at pH 8.0 and that a progressively larger quantity of return sludge increased the uptake rate, by increasing the number of organisms available to accomplish the removal.

In the laboratory, a maximum orthophosphate reduction of 38% was reported when the suspended-solids level was 500 mg/l and the flow rate was 560 cc/min per liter of mixed liquor during a 2-hr aeration period (9 ft^3/gal) (Feng 1962). Another maximum of about 25% was reported when the suspended solids level was 1500 mg/l, the aeration period was 4 hr, and the rate was 280 cc/min (9 ft^3/gal).

Unknown variables such as nutrients in trace quantities, changes in the ecological balance of the microorganisms in activated sludge, and variations in carbohydrate levels may affect profoundly the uptake of various phosphate forms in biological treatment plants. Only recently, the concentrations of phosphorus in the influent and effluent of many treatment plants have been determined. At the Cleveland Easterly Pollution Control Center the average daily total phosphate reduction was 44.4% during December 1965 and 49.8% during February 1966 (Wirtz 1966).

Activated Sludge-Nitrogen Removal

The percent of total nitrogen removed during the modified activated-sludge process, in general, is affected by the degree of active nitrification.

In existing treatment plants, wastewater nitrogen may be converted to nitrate with associated denitrification. Controlled amounts of mixed liquor are recycled from the effluent to the influent zone of aeration. Reduced aeration in the influent zone causes a reduced level of oxidized nitrogen in the recycled mixed liquor to satisfy the high oxygen demand of feed and solids. The gaseous nitrogen is blown out by aeration.

A similar nitrification–denitrification sequence was developed in the laboratory by Ludzack and Ettinger (1962). Their sequence can be applied to activated-sludge plants having operational and, in some cases, plant modifications. The aeration in the influent zone was maintained at a level sufficient to keep the solids in suspension. The dissolved oxygen level reached 0.5 mg/l; this level may have inhibited the complete denitrification in the influent zone. Twenty to 77% of the total nitrogen was removed. This process may be available as a method for nitrogen removal after additional studies have been conducted to improve the controllability.

In pilot-scale laboratory experiments, 90% of the nitrogen forms in the effluent were converted to nitrates during the denitrification step with the following conditions (Wuhrmann 1964):

a) BOD of the raw sewage was 110-130 ppm
b) loads were .3-.5 lb. BOD per lb of organic solids per day, and
c) temperature of the mixed liquor was 14-17°C

Similar percentages of nitrogen were converted when the dissolved oxygen was 1 ppm and 7 ppm in the mixed liquor. The optimum removal of inorganic forms occurred when there was a complete absence of dissolved oxygen. Dissolved oxygen concentrations as low as 0.2 to 0.4 ppm decreased the removal efficiency. The denitrification experiments involved the use of an anaerobic basin placed between the aerator and the secondary clarifier. The detention time of 2.8 hr in the anaerobic tank was based on four critical factors: respiration rate, oxygen concentration, sludge concentration, and denitrification rate of the mixed liquor. The suspended solids in the mixed liquor was 5350 ppm and the dissolved oxygen in the aerator was 4.1 ppm.

A separate pilot-plant experiment was conducted by Wuhrmann (1964) to determine the total nitrogen removals of all nitrogen forms and to compare them with conventional treatment processes. Typical total nitrogen removals by the conventional plant were 40 to 50% (12 to 15 ppm in the effluent); the total nitrogen removals by the denitrification pilot plant, run in parallel with the same influent sewage, were 80 to 90% (3 to 4 ppm in the effluent). The detention times in the aerators and anaerobic basin were 1.67 hr and 2.22 hr, respectively. Remaining operating conditions were:

Denitrification		*Conventional*
5210	MLSS	2050
17.1	ML Temp. °C	17.1
3.8	O_2 in Aerator	1.1

Johnson (1958) reported that seven municipal sewage treatment plants remove 30 to 70% of the total nitrogen. In laboratory-scale experiments on nitrification-denitrification, the efficiencies for removing total nitrogen were 63.3 to 69.2% (Johnson and Schroepfer 1964). Complete balances of all nitrogen forms were reported. The tests were conducted at 20°C and with continuous flow conditions. The laboratory included an anaerobic activated sludge plant, a mechanically mixed reaction compartment followed by a separation zone. Detention times varied from 1.06 to 4.67 hr.

The total nitrogen removal from effluents of conventional, municipal, primary (screening and sedimentation) and secondary (trickling filter or activated

sludge) treatment is about 50% (Stephan 1965b; Keefer 1940). Wuhrmann (1964) found that 48% of the total nitrogen was removed through a trickling filter at low hydraulic loads. This removal corresponded to about 55% nitrification of the effluent. The conditions were: filter depth, 3.5 meters; no recirculation; temperature 14-17°C.

The nitrogen removal efficiencies for conventional treatment processes applied to meat-packing-house effluents, having total organic nitrogen concentrations three times higher than typical municipal wastes, have been found to be (U.S. Department of Health, Education, and Welfare 1965):

Treatment Process	*Total % Nitrogen Removal*
Screening	0
Imhoff tank	10
Trickling filter	45
Activated sludge	70

Algal Systems for Nutrient Removal

At least five important operational characteristics are associated with algal systems to remove nitrogen and phosphorus compounds: 1) high intensity of incident sunlight is desirable to increase photosynthesis, 2) the inorganic carbon supply is rapidly exhausted and may have to be supplemented, 3) temperature fluctuations affect removal efficiences, 4) maintenance of certain algal forms may result in optimum removal, and 5) algal harvesting techniques have often been a limiting factor in the effective removal of nitrogen and phosphorus.

Laboratory studies of three algal species showed that the soluble phosphate content could be reduced by 90% in a high-rate algal culture (Bogan 1961). Contact times were about 6 to 12 hr; nitrogen removals were not reported.

Rand and Nemerow (1964) studied four treatment plants in New York. The Fayetteville, N.Y., subdivision plant at Brookside used trickling filtration followed by oxidation ponds as tertiary treatment. Analyses of the influent and effluent of this plant showed an overall total phosphate removal of 41%. In one run, the total nitrogen was increased by about 38%. However, on the average, almost all of the phosphate removal was attributed to the oxidation pond unit process.

Laboratory studies at Syracuse University by Rand and Nemerow (1965) tested phosphate removal efficiency by controlled algal culture. A 35-day detention period was necessary to remove about 87% of the total phosphate. Even after this period followed by filtration, the filtrate still contained sufficient nutrient material to support algal regrowth. In a separate experiment, 95% of the total phosphate was removed after 37 days detention period (filtered sample). Algae regrowth was observed again. In the latter experiment, approximately 44% was removed after 4 days retention. The light intensities were 200-300 foot candles and the temperatures ranged from 20 to 28°C. The combination of long detention periods and incomplete phosphate removals resulting from these experiments cast doubt on the effectiveness of this process.

Some work has been done on combined biological-chemical systems. Relationships between the concentrations of chemicals necessary to coagulate and precipitate both activated-sludge organisms and phosphate were determined by Tenney and Stumm (1965). With concentrations of alum between 300 and 350 mg/l, more than 97% of the acid soluble phosphate was removed from both settled and filtered activated-sludge effluents. Both phosphates and microorganisms coagulate most effectively when the pH is between 5 to 6.

Studies of the lime treatment of *raw* sewage showed that 80 to 90% of the

total phosphorus and 92 to 98% of soluble inorganic forms were removed (Sawyer and Buzzell 1962); the lower values were observed with recarbonated effluents. The required lime dose was predicted in terms of alkalinity and was independent of the phosphorus content. Six typical raw sewages with alkalinity (as $CaCO_3$) from 75 to 325 mg/l required approximately 100 to 500 mg/l of lime, as $Ca(OH)_2$, to attain a pH of 11.0. It was pointed out that the optimum pH should be determined in each case. About 25% of total nitrogen was removed. The volume of sludge produced was approximately 1% of the volume of treated sewage. Further second-stage polishing of the lime-treated effluent with algal stabilization ponds removed 75 to 80% of the remaining phosphorus compounds, increasing the total phosphorus removal by the two removal processes in sequence, to about 97%. Retention periods were 14 days and 45 days; average total phosphorus removals through the ponds *alone* averaged about 60 and 85%, respectively. Lime treatment was performed on a batch basis. The stabilization pond models were batch-fed once daily.

Chemical Processes

From laboratory studies, Rohlich (1961) asserted that 96 to 99% of soluble phosphates could be removed by using alum, ferrous sulfate, ferric sulfate, or copper sulfate. The use of copper sulfate formed a floc which had poor settling characteristics. The use of alum did not coagulate the inorganic nitrogen compounds, but the soluble phosphates and about 68% of organic nitrogen were removed with about a 200-mg/l dosage of alum. Pilot plant studies, using effluent from Madison Nine Springs sewage treatment plant, showed that 77 to 89% of the soluble phosphates can be removed. Further filtering of the effluent resulted in a total removal of 93 to 97% of the soluble phosphate. Improved settling facilities could result in removal percentages in the range indicated.

Laboratory studies have shown that lime and alum may be the first and second choices for chemical coagulation when considering chemical costs alone (Malhotra, Lee and Rohlich 1964). A lime [$Ca(OH_2)$] dosage of 600 ppm resulted in 90% removal of total phosphorus and an effluent pH of 11.0. A lime dosage of 400 ppm resulted in only 10% removal of total phosphorus and an effluent pH of 10.0. The percent removal of phosphorus was independent of the initial concentration when it ranged from 7 to 35 mg/l of P. The optimum pH for alum flocculation in concentrated activated-sludge effluent was 5.75 ± 0.25. Seventy-eight percent of the total phosphorus can be removed by an alum dosage of 250 ppm at a pH of 8.0; at a pH of 6.0, the same dose may remove 94%. In general, at any alum dosage, the percent removals of orthophosphate and total phosphate are approximately equal.

Other Processes

The proper operation of activated-sludge or high-rate trickling filter plants can minimize nitrification which may result in an effluent amenable to air-ammonia stripping of the effluent. Studies at the University of Wisconsin resulted in ammonia removals of about 78% at air/liquid loadings of 447 cfm/gpm (cubic feet per gallon) (Rohlich 1961). These experimental studies were conducted in 8-inch diameter columns packed with Raschig rings at depths of 2.5 to 7 ft. At the optimum pH of 11, as high as 92.3% of the ammonia nitrogen was removed at about 549 cfm/gpm (cubic feet per gallon) air/liquid loadings. Similar results for nitrogen removal were reported by Nesselson (1954).

The cost of ammonia stripping is likely to be highly variable depending on air requirements. Culp and Slechta (1966) conducted laboratory and pilot-plant studies at the South Tahoe Public Utility District Wastewater Reclamation

Plant. At the South Tahoe plant, NH_3-nitrogen was removed in an air-stripping tower; this corresponded to a 92% removal of all nitrogen in the effluent. They reported that almost all of the ammonia nitrogen may be removed by a full-scale ion exchange process.

Several other processes to remove inorganic nutrients are being tested in the laboratory and at pilot-scale plants, some through assistance from the Federal Water Pollution Control Administration. These are flotation or foam separation, adsorption on activated carbon, electrodialysis, evaporation (distillation), ion exchange, and reverse osmosis.

Stephan (1965b) summarized the capabilities of several advanced treatment methods to remove nitrogen and phosphorus; they are presented in Table 7. By combining conventional plus evaporation processes in series, 96% of total phosphates and 75% of total nitrogen can be removed. Nesselson (1954) found that a high percentage of the anions (including phosphate and nitrate forms) could be removed by using a strongly basic anion exchange resin, but the regenerant volumes equalled at least 7% of the original volume of sewage effluent. The laboratory experiments were conducted on trickling filter effluent. When removing ammonia from an aeration tank effluent with a cation exchange resin, the regenerant was at least 6% by volume of the original total flow of treated sewage. It would be desirable to recover ammonia from the relatively large quantity of residue of the cation exchange process.

As high as 95% of the total phosphate (as P) and 84% of the total nitrogen were removed during laboratory and pilot-scale evaluations of the ion exchange process (Eliassen, Wyckoff and Tonkin 1965) on diatomaceous-earth-filtered activated-sludge effluent. The cost and regeneration potential of the exchange medium were also analyzed. The pilot-scale Water Reclamation Demonstration Plant at Stanford University consisted of secondary activated sludge followed by lime-alum coagulation and sedimentation, diatomaceous earth, and/or sand filtration and ion exchange at the 10 gpm scale. The lime-alum coagulation stage could be omitted. The diatomaceous earth-ion exchange sequence cost about 21 cents per 1000 gal at the 3 MGD scale, including capital cost amortized for 20 years at 3-1/2% assumed interest rate. Of those tested, a strongly basic anion exchange resin was the most satisfactory.

Foster and Ward (1965) summarized the work on intermittent spraying of activated-sludge effluent on the land near Lake Tahoe, Calif. Sewage disposal systems at Tahoe have used land disposal by spraying, and generally it has been effective in protecting public health. The area used was an 80-acre, sparsely forested hillside with an average slope of about 10%. The recently deposited alkali soil was 3 to 40 ft thick. Removal efficiencies of the soil system varied considerably with loading and climatological conditions. Total phosphate removal efficiencies varied from 76 to 93% and total nitrogen from 54 to 68%. However, increased population in the area overburdened the land disposal capability (Ludwig, Kazmierczak and Carter 1964). It should be pointed out that this method of disposal does not prevent nutrients from reaching the lakes by surface runoff or percolation into the ground water system.

Weibel et al. (1966) found the following concentrations of constituents in runoff water from a cultivated 1.45 acre field supporting a winter wheat growth:

Suspended solids	313 mg/l
Chemical oxygen demand	79 mg/l
Biochemical oxygen demand	7 mg/l
Organic nitrogen	9 mg/l
Inorganic (NH_3,NO_2,NO_3)-N	5 mg/l
Total hydrolyzable PO_4	1.7 mg/l

SUMMARY AND CONCLUSIONS

To protect our nation's water resources for multiple use and reuse, the rapid and extensive quality deterioration of the water courses, manifested by accelerated eutrophication, must be stopped. Accelerated eutrophication may be defined as the state of a body of water resulting from the intentional or unintentional, natural or man-made modifications to the aquatic environment to the extent that the ecological system supports an imbalance in the biological production and creates a nuisance or interference with a water use. Through our research effort, we must be able to develop practical control schemes; they may or may not be based on the techniques and concepts which at this time seem to be the most promising.

An attempt to define the problem and its magnitude only "scratches the surface" but it does reveal specific areas where research is desperately needed. The nutrient and micronutrient requirements of algae to produce nuisance blooms and the ecological kinetics of the aquatic environment for all life forms must be investigated further. Treatment processes and techniques must be developed to incorporate control methodology, and new analytical measurement techniques must be developed, perfected and used. Point-source treatment processes such as electrodialysis, chemical treatment, ion exchange, and distillation to remove nutrients must be evaluated in pilot-scale and full-scale plants. Non-point-source control techniques such as the mining of bottom deposits, algal harvesting and in-stream treatment of tributaries should be investigated further. Therefore, the questions that remain are not whether research on these techniques should be done, but instead how much of a research effort must be initiated in the very near future; which techniques will be the most effective and practical; and when will they be available for use in stopping a threat to our water supplies—a basic necessity for life.

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